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Los Alamos
NATIONAL LABORATORY

DANCE
Device for Measurement of (n,γ) Reactions on Radioactive Species

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DANCE (Device for Advanced Neutron Capture Experiments) is a 4π 162 element BaF_2 array under development at Los Alamos National Laboratory. It is designed to provide high granularity, fast timing and high photon detection efficiency. It will be located at the Los Alamos Neutron Scattering Center where neutrons are produced using 800 MeV proton induced spallation reactions on heavy element production targets. Using the pulsed high neutron fluence available at this facility combined with time of flight techniques it will be possible to make neutron capture measurements in the neutron energy range from eV to 100's of keV on rare and radioactive target material at the milligram and below level. These measurements will provide critically needed data for the interpretation of the astrophysical s-process "branching point" nuclei as well as information for reactions needed in understanding transmutation processes of radioactive species.

I. Introduction

One of the major scientific questions has dealt with the origin of the elements. The modern understanding of heavy element nucleosynthesis can be traced to the pioneering work in the 1950's of Cameron[1)] and that of Burbidge, Burbidge, Fowler and Hoyle (BBFH).[2)] The major modes of production are through a rapid (or "r" process) and a slow (or "s" process) with some weaker processes making measurable contributions. Though these breakthrough works have served as very valuable guidelines there has been substantial progress made in the ensuing years.[3,4)] The r-process occurs in the very high neutron fluence region generated in the explosive environment of supernova and since it involves nuclear reactions on isotopes far removed from the line of beta stability it can be difficult to quantitatively interpret yields. On the other hand, the s-process, which occurs in the asymptotic giant branch (AGB) of low to medium mass stars (or in red giants), always remains near the line of stability since the capture rate is slow compared with most beta decay rates. For the s-process the general trends of isotopic production are well reproduced by modeling.[5,6)] For a class of nuclei the stellar neutron capture rate and their beta decay lifetimes are comparable. These so called "branching point" nuclei are of high interest since a detailed understanding of this competition provides insight into the dynamics of the production process. By observing the isotopic yields the stellar neutron fluences can be deduced if the neutron capture cross sections are known for the isotopes. By definition these branching point nuclei are unstable, which means cross sectional information on such species is required. In general, very little is known experimentally for reactions on these isotopes and, therefore, most predictions of stellar neutron dynamics are based on theoretical modeling of the underlying microscopic nuclear reaction set.

In order to experimentally provide data in this critical region we are developing a new detector that will be capable of measuring neutron capture events from the eV through the 100's of keV region on very small quantities of radioactive materials. The emphasis of this program will be to study reactions on unstable species for which little or no experimental data exist. Knowledge of reaction probabilities on these species is of high importance in fields ranging from nucleosynthesis to transmutation of radioactive wastes. The possibilities of doing these measurements require a confluence of capabilities beginning with intense neutron sources, production and isolation of the desired isotopes and highly efficient detector arrays for studying the reactions.

Spallation driven neutron sources[3,6)] deliver white neutron beams and thus are efficient methods for measuring neutron excitation functions using time of flight techniques. Another crucial aspect is having the capability to produce, purify and prepare suitable target materials of exotic species. This involves methods to produce isotopes in near mg quantities and can often be accomplished through high current spallation reactions on thick targets or selected irradiations of suitable materials in high flux nuclear reactors. Of course, such methods lead to products that are often highly radioactive. Thus suitable processing facilities are required that can work with materials that are often at the multi Curie level of activity. Since many of the producing reaction techniques are not very specific, radiochemistry and often mass separation are needed to make target materials of suitable purity. Finally a detection system has to be built that is highly efficient for working with the relatively low neutron beam fluence and the small amount of target material. The detector has to also be able to function in the high radiation field generated by the radioactivity of the target as well as having methods for discriminating against effects of the inevitable scattered neutron beam. All of these capabilities are discussed below, with special emphasis placed on the development of the detector array - DANCE.

II. Basic Properties

Neutron capture (Fig. 1) is a process that transforms an isotope (^{A}Z) into the next heavier isotope (^{A+1}Z). For direct measurement of the reaction we utilize the fact that the capture mechanism results in excitation energy in the heavier isotope corresponding to the kinetic energy of the incident neutron plus the neutron separation energy for the isotope. These separation energies for nuclei near stability are typically in the 6-8 MeV range. This energy is liberated via electromagnetic transitions, usually in the form of cascading gamma rays. The key ingredients that are used to identify and characterize the reactions are: the deexcitations primarily occur promptly after capture, they have a relatively high total energy release, and have a multiplicity of transitions. For s-process nucleosynthesis a complex network

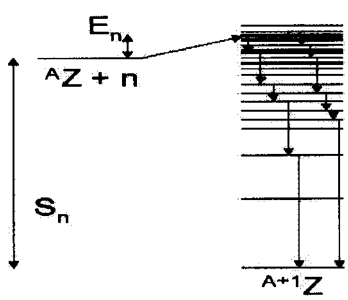

Fig. 1 Neutron Capture Systematics.

of reactions occur to build up the population of the heavier elements. Fig. 2 shows the reaction sequence in the region of the Sm isotopes. In the stellar neutron environment the stable nuclei can only undergo capture reactions to make heavier isotopes. However, once the capture process results in a beta unstable nucleus then there becomes the possibility for a competition between capturing another neutron to make yet a heavier isotope or to beta decay to form a heavier element. For a beta decay constant - λ, an average stellar neutron fluence - ϕ, and a neutron capture cross section - σ, then if:

$$\lambda/\phi\sigma >> 1; \rightarrow \text{Beta Decay}$$
$$\lambda/\phi\sigma << 1; \rightarrow \text{Neutron Capture}$$
$$\lambda/\phi\sigma \sim 1; \rightarrow \text{Capture/Beta decay Competition}$$

An isotope of particular interest is ^{152}Sm which has a 93 year half-life. Current predictions[3)] are the stellar nucleosynthesis occurs with a cyclical burning period that consists of an ~20 year pulse of neutrons being generated by the ^{13}C(α,n) reaction with a Maxwellian temperature of ~10 keV followed by an ~1 year burst of neutrons from the ^{22}Ne(α,n) at temperatures nearer to 30 keV (fig 3). This type burning cycle implies that isotopes having half lives in the few year range will be the primary "branching point" nuclei and have the highest sensitivity for determining information regarding the dynamics of the stellar processes. Of course, most of the isotopes that are of highest interest, since they are radioactive, have little or no experimental data associated with the capture process, especially at the neutron energies that are most appropriate for nucleosynthesis.

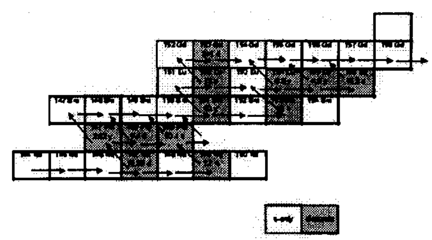

Fig. 2 S-Process Nucleosynthesis in the Sm region.

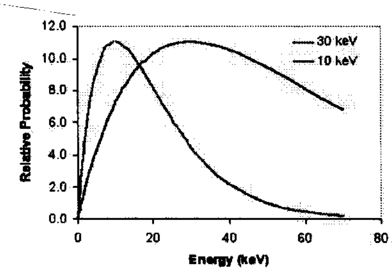

Fig. 3 Maxwellian Distributions for temperatures of 10 keV (^{13}C(α,n)) and 30 keV (^{22}Ne(α,n))

III. Experimental Parameters

1. Neutron Production

The experimental program will be carried out at the Lujan Neutron Scattering Center, which is part of the Los Alamos Neutron Scattering Center (LANSCE) at the Los Alamos National Laboratory. Protons from the 800 MeV LINAC are injected into a Proton Storage Ring and compressed to provide high intensity short pulses. The integrated delivered beam can be up to 200 μA at a rate of 20 pulses/sec each about 250 nsec wide. For more details see.[9] At the Lujan Center there are 16 beam lines radiating around the target/moderator system that is fed by vertically injected protons from the Proton Storage Ring (fig. 4). The DANCE detector array will be deployed on flight path 14 which is a new flight path currently under construction.

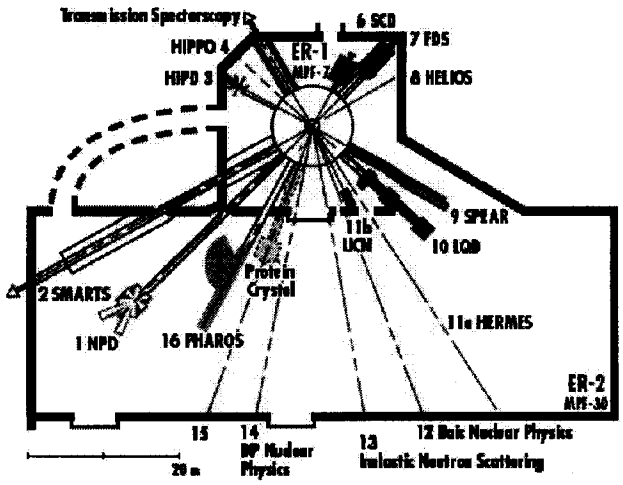

Fig. 4 Plan view of the Los Alamos Lujan Neutron Scattering Center. The DANCE array is on Flight Path 14.

Construction of the flight path is nearing completion and will consist of four discrete collimators each with alternating copper and 5% borated polyethylene layers and having changeable inserts. Each collimator section is 81 cm long consisting of 51 cm of Cu and 30 cm of borated polyethylene. The final collimator ends 19.2 m from the neutron source. The target location within the DANCE detector will be at 20.5 m from the source. The last collimator is 0.6 cm in diameter, which allows a beam spot that is uniform out to r=0.3 cm at the target location and has a flux that falls to 1/100 of the central value by r=0.75 cm.

The calculated on target neutron flux in FP-14 is shown in Fig. 5. The flux is presented in units of neutrons/(eV-pulse) (there are 20 pulses/sec) and is for a primary proton storage ring current of 200 μA.

2. Target Preparation

For these experiments target quantities of around 1 mg are required. Since the half-life region of high interest is

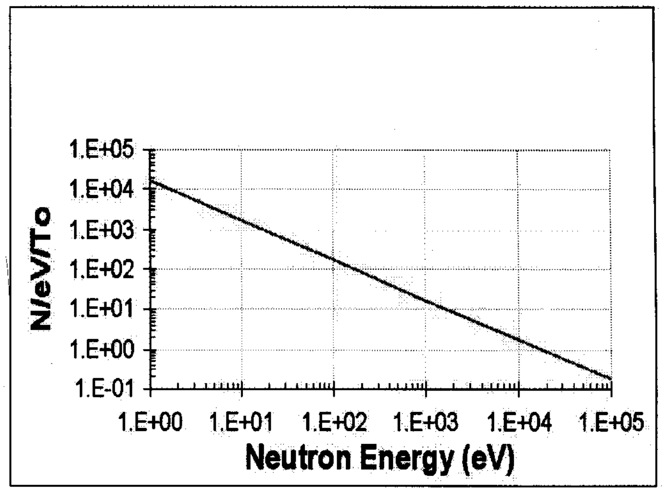

Fig. 5 Calculated neutron flux at the DANCE irradiation position on Flight Path 14.

in the year range this necessitates working with target materials that have radioactivity levels on the order of a Curie. Such high activity level targets require specialized handling capabilities. Target materials can be produced in a variety of ways. At Los Alamos we have had an extensive program for the production of radioactive isotopes for medical applications.[10] These isotopes have been produced by irradiating cm thick targets located near the LINAC beam stop. Such targets have been exposed to upwards of 1 A-hr of 800 MeV protons. Under these conditions spallation reactions can produce multi milligram quantities of many isotopes. Once the irradiation cycle was completed the targets were remotely placed in highly shielded containers and transported to a hot cell chemical facility for processing. The medical isotope program extracted materials of interest to their program, but remaining byproduct isotopes could be made available for other applications.

In addition to spallation reactions, other techniques are possible for production of these macro quantities of isotopes. Irradiation in high flux reactors is often a viable technique to produce isotopes near the line of beta stability. Other techniques could involve the use of lower energy light ion accelerators to produce mostly neutron deficient isotopes.

Though chemical processing facilities exist at Los Alamos for handling these highly radioactive species, quite often the producing reaction techniques do not result in an isotopically pure material. The chemistry can isolate the desired element, but a physical process is often required to obtain the desired material in a sufficiently high purity state to perform the neutron capture experiments. For this purpose we have constructed a dedicated Radioactive Species Isotope Separator (RSIS). It is specifically designed to separate highly radioactive isotopes. The separator shown in Fig. 6 is housed entirely inside of a hot cell. The ion optics of RSIS are designed to allow up to 100 μA operation and to minimize of cancel major aberrations. Test operations of the separator have shown separation factors between adjacent isotopes of about 10^4. The ionization efficiency is element dependent and typically on the order of 5 to 30%. The separator is designed to run in a stable manner to permit largely unattended operation. Collection periods on the order of days are required to obtain sufficient material for the neutron capture studies.

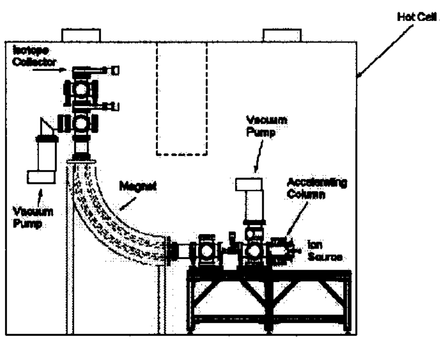

Fig. 6 The Radioactive Species Isotope Separator - RSIS.

3. DANCE Detector

The design objective for the DANCE detector is to be able to measure neutron capture reactions for unstable species. To accomplish this several considerations are important. First the target material is rare and highly radioactive. Thus this requires a highly efficient detector system that will be operable with small amounts of material - i.e., a 4π system with sufficient stopping power to collect all emitted radiation. Second the high radioactivity of the targets drives the system toward having a highly granular array of detection elements so as to minimize background pile up problems associated with any single element. Thirdly, since the gamma transitions following the capture event are emitted promptly it is also highly important to have a system with fast timing so as again to minimize backgrounds from the gamma emissions from the target. Fourth the backgrounds from the target are limited in total energy emission to their Q_β values which are substantially below the neutron binding energy emission that occurs following capture. Therefore, it is desirable to have a detector system that functions as a calorimeter to provide additional discrimination between the desired signal and the target background radiations.

To build a 4π detector with detection elements that each subtend the same solid angle requires the use of specific "magic" numbers of crystals.[11] We have chosen a configuration that has 162 elements consisting of 12 elements of regular pentagons, 30 elements of regular hexagons and 60 each of two irregular shapes of hexagons (Fig. 7). Several types of scintillators are potentially useable for such an array. Some of the properties of these materials are listed in Table 1. We have chosen BaF_2 for our system since it has the widest combination of desirable properties including: high density (and high Z) for efficient stopping of gamma rays, a sub nanosecond decay component to allow fast timing and a reasonable photon production yield to allow adequate resolution. BaF_2 is also non hydroscopic for ease of handling, has a relatively small neutron capture probability and is available commercially. The DANCE crystals are each 15 cm deep and have a front plane that is 17 cm from the center of the beam line.

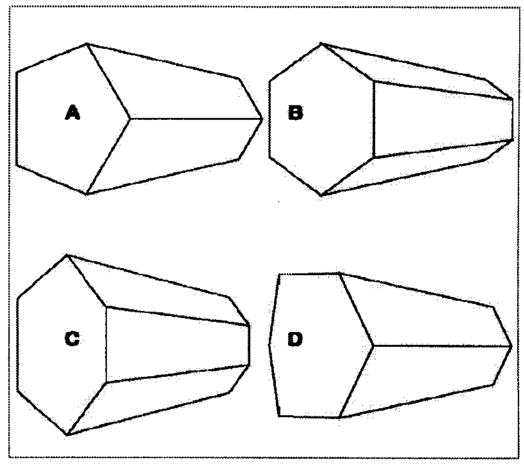

Fig. 7 The 4 DANCE crystal shapes.

Table 1 Scintillator Properties

Material	Density	Decay Time	Wavelength	Photons/MeV
	g/cc	ns	nm	
BaF_2	4.88	0.6, 630	180-310	1,800; 10,000
$Bi_4Ge_3O_{12}$	7.13	60, 300	480	700; 7,500
NaI(Tl)	3.67	230	415	38,000
CsI(Tl)	4.51	1000	540	59,000
CsI(pure)	4.51	8	315	2,300
CeF_3	6.16	3,27	300,340	200; 4,300
C_6D_6	0.954	2.8	425	10,000

Figure 8 is a schematic showing a cut away view of the DANCE array with inserted beam pipe and target/shielding assembly. Figure 9 is a photograph of the DANCE support assembly and Figure 10 is a photograph of a BaF_2 crystal element.

IV. Simulations and Results

We have performed extensive simulations of the parameters associated with the experiment.[12] We have also been able to draw on the experience of the Karlsruhe 42

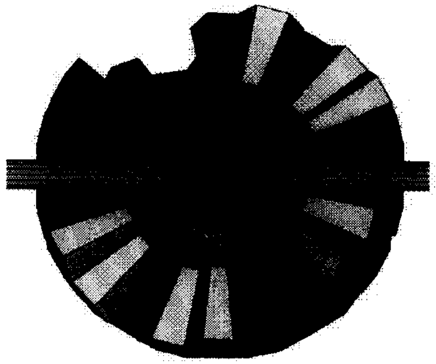

Fig. 8 DANCE detector cut away view.

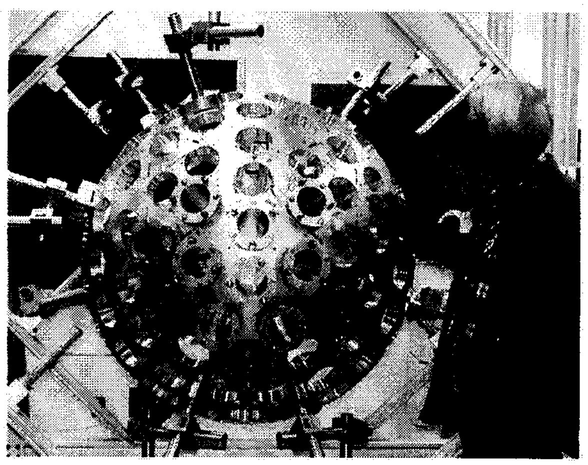

Fig. 9 DANCE crystal support structure.

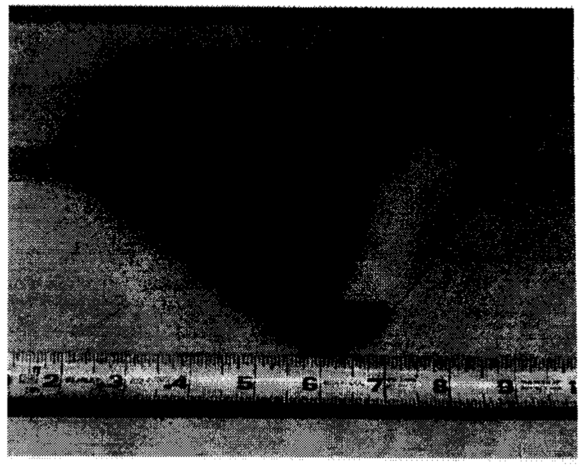

Fig. 10 A BaF_2 crystal for DANCE

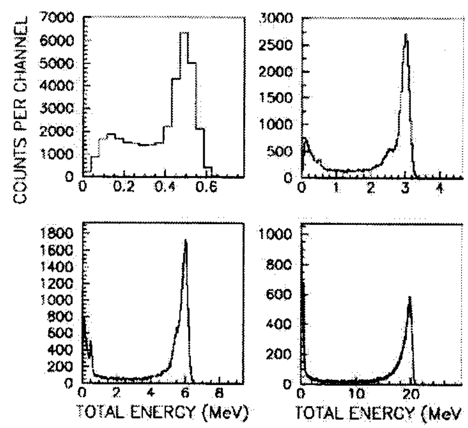

Fig. 11 Monte Carlo calculation of the Energy Deposited in DANCE for 4 incident energies.

element 4π detector.[13] Figure 11 shows the calculated energy deposited in the array for monoenergetic photons of 0.5, 3, 7 and 20 MeV. It is seen that reasonable resolution is obtained with a large fraction of the energy being deposited in the summed photopeak. Figure 12 shows the calculated average multiplicity for the complete set of crystals as a function of the photon energy. Also shown are the "cluster" multiplicity where a cluster is defined as crystals adjacent to the hit crystal. This shows that most of the inelastically scattered events generate signals in adjacent detectors. This property can be used as an additional method to distinguish high multiplicity events following neutron capture from those associated with low multiplicity emission from background events.

An isotope of interest for capture studies is 1.9 yr ^{171}Tm. For a 1 mg/cm2 target irradiated in DANCE on FP-14 we would expect the event rate shown in Fig. 13. The results are shown for a 200 μA proton beam and for a dE/E of 10% neutron energy bin. Since our interest is mostly in energies above the highly fluctuating resonance regions, taking such a large energy interval is acceptable. The capture rates even at neutron energies as high as 100 keV are around 0.1/sec. This is a very acceptable rate and implies that even smaller than mg quantities of target materials may often be possible. To demonstrate the feasibility of working with these small amounts of materials we have performed preliminary experiments on Tm isotopes using a different beam line and much less efficient C_6D_6 detectors.[14] The results for measurements on the stable isotope ^{169}Tm are shown in Fig. 14 and the results for measurements on the ^{171}Tm are presented in Fig. 15. Both targets were approximately 1 mg/cm2 and prepared in a similar fashion. The ^{169}Tm results agree well with the known cross section and the GNASH calculations, while the ^{171}Tm results show a significant deviation from the predictions. We will, of course, repeat this preliminary experiment with the much more appropriate DANCE array when it is operational.

V. Conclusions

The DANCE array is nearing completion. When operational this new capability will offer the possibility of making neutron capture measurements on a whole suite of rare and radioactive isotopes. These cross sections will provide new insights in to astrophysical issues as well as

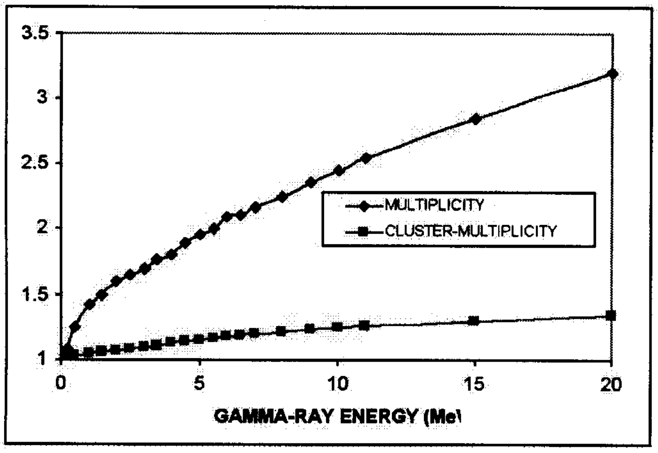

Fig. 12 Monte CArlo calculated multiplicity of single and "cluster" crystal hits as a function on incident photon energy.

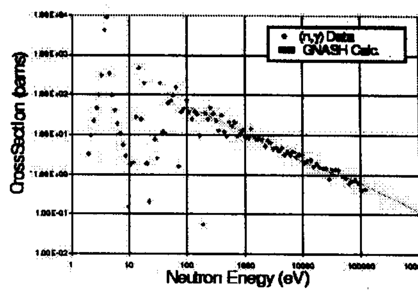

Fig. 14 Measured and model calculated (n, γ) cross section for stable ^{169}Tm

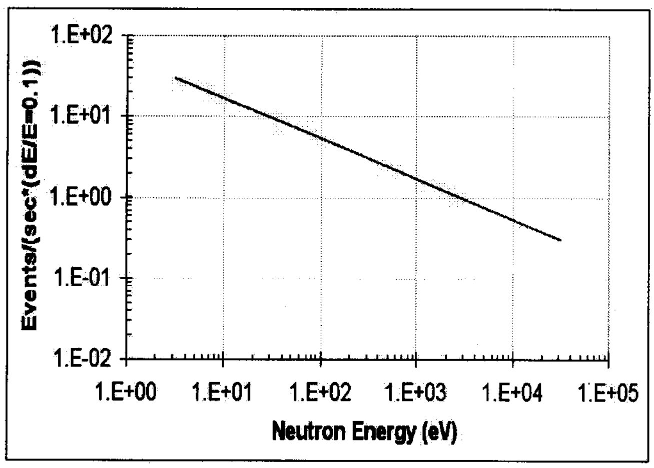

Fig. 13 Calculated event rate for a 1 mg/cm^2 ^{171}Tm target measured with DANCE.

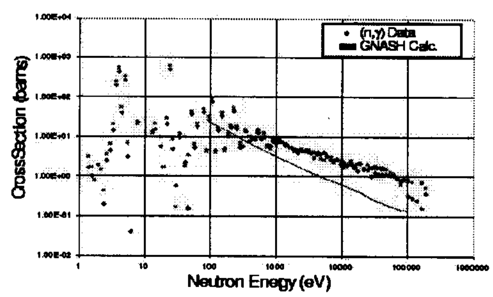

Fig. 15 Preliminary measured and claculated (n, γ) cross sections for the radioactive 1.9 yr ^{171}Tm isotope

complementary programs dealing with the transmutation of radioactive species in high neutron fluence environments.

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